

Strong Suppression of Magnetic Ordering in an $S = 1/2$ Square-Lattice Heisenberg Antiferromagnet $\text{Sr}_2\text{CuTeO}_6$

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The most appealing physical phenomenon in frustrated quantum magnets is the emergence of nonclassical ground states stabilized by the synergy effect of the spin frustration and the quantum fluctuation. The spin-1/2 square-lattice Heisenberg antiferromagnet (SLHAF) with the nearest-neighbor J_1 and next-nearest-neighbor J_2 exchange interactions is a typical frustrated quantum magnet, for which energetic theoretical studies have been performed. Most of the theoretical results suggest that the $S = 1/2$ $J_1 - J_2$ SLHAF exhibits the quantum disordered ground state for $\alpha_{c1} < J_2/J_1 < \alpha_{c2}$ with $\alpha_{c1} \approx 0.4$ and $\alpha_{c2} \approx 0.6$.¹⁻¹¹ However, the nature of the ground state has not been theoretically clarified. The ground states for $\alpha_{c1} > J_2/J_1$ and $\alpha_{c2} < J_2/J_1$ are the Néel antiferromagnetic state and the columnar antiferromagnetic state, respectively.

On the experimental side, many materials have been investigated from the viewpoint of the $S = 1/2$ $J_1 - J_2$ SLHAF.¹²⁻²⁰ However, the quantum disordered ground state has not been observed experimentally. The search for approximate realizations of the $S = 1/2$ $J_1 - J_2$ SLHAF with the critical parameter range has been continued.

In this short note, we report the results of the magnetic and specific heat measurements on a double perovskite, $\text{Sr}_2\text{CuTeO}_6$. This compound crystallizes in the tetragonal structure with the space group $I4/m$, as shown in Fig. 1.^{12,21} The crystal structure consists of CuO_6 and TeO_6 octahedra, shaded blue and maroon, respectively, which are linked sharing their corners. All the CuO_6 octahedra are elongated along the crystallographic c axis owing to the Jahn-Teller effect. Consequently, the hole orbitals $d_{x^2-y^2}$ of Cu^{2+} ions with spin-1/2 are spread in the ab plane, in which Cu^{2+} ions form a uniform square lattice, as shown in Fig. 1(b). This leads to the strong superexchange interaction in the ab plane and the weak superexchange interaction between the ab planes. Thus, it is expected that $\text{Sr}_2\text{CuTeO}_6$ can be magnetically described as a quasi-two-dimensional $S = 1/2$ $J_1 - J_2$ SLHAF [Fig. 1(c)].

Figure 2(a) shows the magnetic susceptibility of $\text{Sr}_2\text{CuTeO}_6$ powder as a function of temperature. Our susceptibility data is consistent with that reported in Ref. 12. With decreasing temperature, the susceptibility exhibits a rounded maximum at $T_{\text{max}} \approx 73$ K and decreases. This behavior is characteristic of two-dimensional SLHAFs,^{18,22,23} and is common to the susceptibilities in A_2CuMO_6 with $\text{A} = \text{Ba}$, Sr and $\text{M} = \text{W}$, Mo , Te .^{12,13,15}

Figure 2(b) shows the low-temperature specific heat in $\text{Sr}_2\text{CuTeO}_6$ measured at zero magnetic field. A sharp λ -like anomaly indicative of the magnetic ordering is observed at

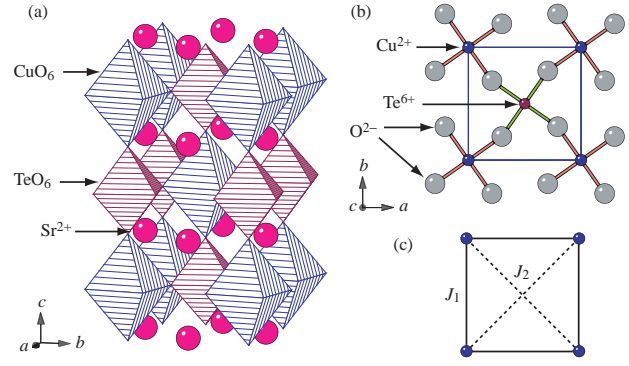


Fig. 1. (Color online) (a) Schematic crystal structure of $\text{Sr}_2\text{CuTeO}_6$. The blue octahedron is a CuO_6 octahedron with a Cu^{2+} ion at the center, and the TeO_6 octahedron is shaded maroon. (b) Crystal structure viewed along the c axis. Magnetic Cu^{2+} ions with spin-1/2 form a uniform square lattice in the c plane. Dotted blue lines denote the chemical unit cell. (c) Exchange interactions J_1 and J_2 in the ab plane.

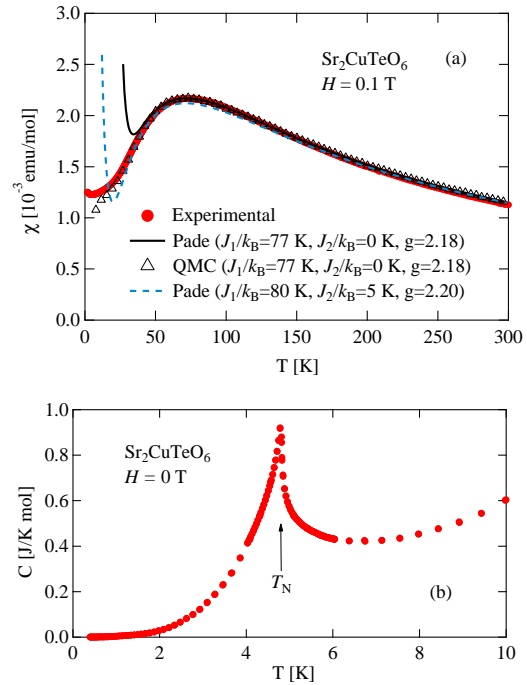


Fig. 2. (Color online) (a) Temperature dependence of magnetic susceptibility χ in $\text{Sr}_2\text{CuTeO}_6$ measured at $H = 0.1$ T (circular symbols). The solid and dashed lines show the susceptibilities calculated by the Padé approximation with $(J_1/k_B, J_2/k_B, g) = (77 \text{ K}, 0 \text{ K}, 2.18)$ and $(80 \text{ K}, 5 \text{ K}, 2.20)$, respectively. The susceptibility calculated by the quantum Monte Carlo (QMC) method with $(J_1/k_B, J_2/k_B, g) = (77 \text{ K}, 0 \text{ K}, 2.18)$ is shown by triangular symbols. (b) Low-temperature specific heat in $\text{Sr}_2\text{CuTeO}_6$ measured at zero magnetic field.

$T_N = 4.8$ K. This Néel temperature is much lower than those observed in isostructural A_2CuMO_6 with $\text{A} = \text{Ba}$, Sr and $\text{M} = \text{W}$, Mo , in which $T_N = 24 - 28$ K.^{14,15} The strong suppression of magnetic ordering in $\text{Sr}_2\text{CuTeO}_6$ should be ascribed to the weakness of the interlayer exchange interaction or to the strong spin frustration in the square lattice.

Before estimating the exchange constants from the susceptibility data, we discuss the superexchange interactions J_1 and J_2 in terms of the Kanamori theory.²⁴ In the isostructural Ba_2CuWO_6 , the antiferromagnetic J_2 interaction is dominant and the next-nearest-neighbor spins form the antiferromagnetic state below $T_N = 28$ K.¹⁴ This finding suggests that

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the exchange path $\text{Cu}^{2+} - \text{O}^{2-} - \text{M}^{6+} - \text{O}^{2-} - \text{Cu}^{2+}$ is dominant in A_2CuMO_6 . Figure 3 illustrates the orbital configurations in $\text{Sr}_2\text{CuTeO}_6$. For simplification, we assume that $\text{Cu}^{2+} - \text{O}^{2-} - \text{M}^{6+}$ is a straight line, although it is actually a zigzag line, as shown in Fig. 1(b). The sign of the superexchange interaction strongly depends on the filled outermost orbital, which is the $4d$ orbital for $\text{Sr}_2\text{CuTeO}_6$. We consider the superexchange interaction between hole spins on the $d_{x^2-y^2}$ orbitals of Cu^{2+} ions. The superexchange interaction J_1 between $\text{Cu}^{2+}(1)$ and $\text{Cu}^{2+}(2)$ is based on the following perturbation process. (i) Hole 1 with up spin on $\text{Cu}^{2+}(1)$ is first transferred to the p_x orbital of O^{2-} , which is combined with the $d_{x^2-y^2}$ orbital of Te^{6+} to form a molecular orbital. (ii) Hole 2 on $\text{Cu}^{2+}(2)$ is also transferred to other molecular orbital composed of the p_y orbital of O^{2-} and the $d_{x^2-y^2}$ orbital of Te^{6+} . In this case, the two hole spins on the same $d_{x^2-y^2}$ orbital of Te^{6+} must be antiparallel owing to the Pauli principle. (iii) The two holes are transferred back to the $d_{x^2-y^2}$ orbitals of two Cu^{2+} ions. Consequently, an antiferromagnetic superexchange interaction takes place between $\text{Cu}^{2+}(1)$ and $\text{Cu}^{2+}(2)$. A similar perturbation process is also applicable to the superexchange interaction between $\text{Cu}^{2+}(1)$ and $\text{Cu}^{2+}(3)$. Thus, J_2 becomes antiferromagnetic. There are two $\text{Cu}^{2+} - \text{O}^{2-} - \text{Te}^{6+} - \text{O}^{2-} - \text{Cu}^{2+}$ paths for J_1 , whereas for J_2 , the exchange path is single. The contributions of the paths to the superexchange interaction should be almost the same. Thus, if the other exchange paths are negligible, the condition $J_2/J_1 \approx 1/2$, which is in the critical region of the quantum disordered ground state, should be realized in $\text{Sr}_2\text{CuTeO}_6$. On the other hand, for $\text{M} = \text{W}$ and Mo , the p_x and p_y orbitals of M^{6+} are orthogonal to each other (inset of Fig. 3). Hence, two hole spins 1 and 2 on the p_x and p_y orbitals, respectively, must be parallel owing to the Hund rule. This leads to the ferromagnetic superexchange interaction J_1 between $\text{Cu}^{2+}(1)$ and $\text{Cu}^{2+}(2)$, while J_2 becomes antiferromagnetic as in the case of $\text{M} = \text{Te}$.

Here, we estimate the exchange parameters J_1 and J_2 of $\text{Sr}_2\text{CuTeO}_6$ from the susceptibility data by the [5, 5] Padé approximation using the result of high-temperature expansion up to tenth-order of $\beta = 1/k_B T$.¹⁸⁾ We also calculate the susceptibility for $J_2 = 0$ case by the quantum Monte Carlo (QMC) method for the 12×12 site square cluster.²⁵⁾

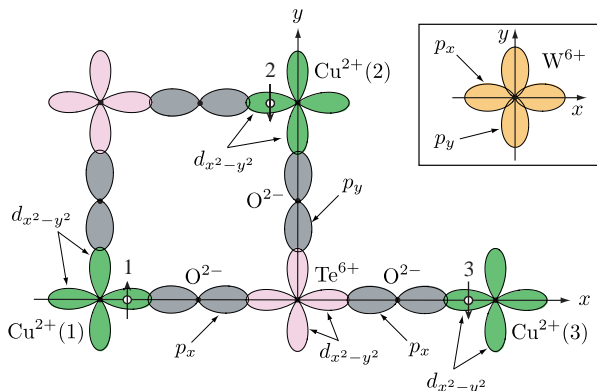


Fig. 3. (Color online) Illustrations of orbital configurations related to superexchange interactions in A_2CuMO_6 through $\text{Cu}^{2+} - \text{O}^{2-} - \text{M}^{6+} - \text{O}^{2-} - \text{Cu}^{2+}$ for $\text{M} = \text{Te}$. The inset shows the configurations of the filled outermost p orbitals in $\text{M} = \text{W}$ and Mo .

From the above discussion on the superexchange interactions, we can deduce that J_1 and J_2 interactions are both antiferromagnetic and $J_1 > J_2$. We calculated susceptibilities varying J_2/J_1 , and compared the results to the experimental susceptibility. It was found that J_1 is much larger than J_2 in $\text{Sr}_2\text{CuTeO}_6$ in contrast to the case of $\text{M} = \text{W}$ and Mo . We show the susceptibilities calculated by the Padé approximation with $(J_1/k_B, J_2/k_B, g) = (77 \text{ K}, 0 \text{ K}, 2.18)$ and $(80 \text{ K}, 5 \text{ K}, 2.20)$, respectively, and that calculated by the QMC method with $(J_1/k_B, J_2/k_B, g) = (77 \text{ K}, 0 \text{ K}, 2.18)$, which coincides with the result obtained by the Padé approximation for $T > 40 \text{ K}$. As shown by the solid line and triangular symbols in Fig. 2(a), the susceptibility is approximately represented in terms of a simple square-lattice antiferromagnetic Heisenberg model ($J/k_B = 77 \text{ K}$) without the next-nearest-neighbor interaction J_2 . From the analysis of the susceptibility, exchange parameters in $\text{Sr}_2\text{CuTeO}_6$ are estimated as $J_1/k_B \approx 80 \text{ K}$ and $J_2/J_1 < 0.07$. This result indicates that a superexchange path other than $\text{Cu}^{2+} - \text{O}^{2-} - \text{Te}^{6+} - \text{O}^{2-} - \text{Cu}^{2+}$ path, e.g., $\text{Cu}^{2+} - \text{O}^{2-} - \text{O}^{2-} - \text{Cu}^{2+}$ path, also makes an important contribution to J_1 interaction. For the accurate evaluation of the exchange parameters in $\text{Sr}_2\text{CuTeO}_6$, further experiments, such as a dispersion measurement, are necessary.

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